# INTEGRATED REACTORS, METHODS OF MAKING SAME, AND METHODS OF CONDUCTING SIMULTANEOUS EXOTHERMIC AND ENDOTHERMIC REACTIONS

## RELATED APPLICATIONS

This application is a continuation of U.S. patent application Ser. No. 11/232,485, now U.S. Pat. No. 7,803,325, which is a divisional of U.S. patent application Ser. No. 10/076,875, now U.S. Pat. No. 6,969,506, which was a continuation-in-part of U.S. patent applications Ser. No. 09/375,614, now U.S. Pat. No. 6,488,838 and Ser. No. 09/640,903, now U.S. Pat. No. 6,680,044, which are incorporated herein as if reproduced in full below. In accordance with 35 U.S.C. sect. 119 (e), this application claims priority to U.S. Provisional Application No. 60/269,628, filed Feb. 16, 2001.

## FIELD OF THE INVENTION

The present invention relates to integrated reactors for conducting exothermic and endothermic reactions, methods of making integrated reactors, and methods of conducting reactions in integrated reactors.

## INTRODUCTION

Currently, endothermic reactions performed in microreactors are driven using heat from an external source, such as the effluent from an external combustor. In doing so, the temperature of the gas stream providing the heat is limited by constraints imposed by the materials of construction. For example, a typical microreactor constructed from Inconel 625 might be limited in use for gas service to temperatures of ~1050° C. or less. Practically, this means that the effluent from an external combustor must be diluted with cool gas (i.e. excess air) to bring the gas temperature down to meet material temperature constraints. This increases the total gas flow rate, raising blower/compressor costs. Moreover, heating the gas stream externally introduces heat losses (associated with delivery of the hot gas to the microreactor) and expensive high temperature materials to connect the combustor to the microreactor.

On the other hand, an integrated combustor can produce heat for the reaction in close proximity to the reaction zone, thus reducing heat losses and increasing efficiency. Because traditional combustion catalysts are not stable at high temperatures (above ~1200° C.) due to noble metal sintering, the integrated combustor must remove heat at a rate sufficient to keep local temperatures at the catalyst surface below this level or risk rapid catalyst deactivation.

## SUMMARY OF THE INVENTION

In an integrated reactor, combustion/heat generation should occur in close proximity to the endothermic reaction. Preferably, an exothermic reaction occurs in microchannels that are interleaved with microchannels in which there is an endothermic reaction. Co-flow of endothermic and exothermic reaction streams is preferred; however, cross-flow or countercurrent flow is also an option. The heat of an exothermic reaction is conducted from the exothermic reaction catalyst to the endothermic reaction catalyst, where it drives the endothermic reaction. This rapid heat removal from the combustion region enables the option to use a very small fraction of excess air (e.g., close to stoichiometric combustion, which could produce temperatures exceeding 1400° C. if reacted adiabatically). The use of a flow-by catalyst configuration for one or both the exothermic and endothermic microchannels can create an advantageous capacity/pressure drop relationship. In a flow-by catalyst configuration, gas flows in a 0.05-2 mm gap adjacent to a thin layer of engineered catalyst that contacts the microchannel wall. The catalyst may be either inserted and adjacent to the reactor wall or integral with the reactor wall. In the case of integral with the reactor wall, a preferred method is washcoating a catalyst on a wall or walls of the microchannel The catalyst may include the use of additional layers for increasing surface area, such as a porous high surface area ceramic, or layers for promoting adhesion of a ceramic to metal, such as amorphous titania that is either CVD or solution deposited. The use of channels having a minimum dimension of more than 2 mm may be less effective since heat and mass transfer limitations may be magnified. An integrated combustor can use the high surface area of microreactor channels to remove heat as it is produced, thus keeping microreactor components from exceeding material temperature constraints while combusting with much less excess air (or diluent) than would be necessary for an external combustor.

In one aspect, the invention provides an integrated reactor, that includes: a first reaction chamber having a width of 2 mm or less, where there is an open channel through the first reaction chamber, wherein the first reaction chamber has an internal volume comprising 5 to 95 vol. % of porous catalyst and 5 to 95 vol. % of open space. The integrated reactor also includes a second reaction chamber having a width of 2 mm or less, wherein there is an open channel through the second reaction chamber, wherein the second reaction chamber has an internal volume comprising a catalyst and at least 5 vol. % of open space; and a reaction chamber wall separating the first chamber and the second chamber. This intergrated reactor possesses a heat flux characteristic of at least 1 W/cc as measured according to the Heat Flux Measurement Test.

The invention also includes methods of performing exothermic and endothermic reactions in the reactor. An exothermic reaction composition is a chemical composition that will react under the selected conditions to produce heat; typically a catalyst will catalyze the reaction.

In another aspect, the invention provides an integrated reactor, that includes: a first reaction chamber having a width of 2 mm or less, wherein there is an open channel through the first reaction chamber, wherein the first reaction chamber has an internal volume comprising 5 to 95 vol. % of porous catalyst and 5 to 95 vol. % of open space; and a second reaction chamber having a width of 2 mm or less, wherein there is an open channel through the second reaction chamber, wherein the second reaction chamber has an internal volume comprising a catalyst and at least 5 vol. % of open space. A reaction chamber wall separates the first chamber and the second chamber; and the intergrated reactor possesses a $NO_x$ output characteristic of less than 100 ppm as measured according to the Standard $NO_x$ Test Measurement.

The invention also provides a method of making an integrated reactor, comprising: providing a single block of thermally conductive material; forming at least one first microchannel in the block; forming at least one second microchannel in the block; placing at least one catalyst capable of catalyzing an exothermic reaction in the at least one first microchannel; and placing at least one catalyst capable of catalyzing an endothermic reaction in the at least one second microchannel. In the integrated reactor, the first microchannel and second microchannel are separated by less than 1 cm.